Title: **DEVELOPMENT OF HIGHLY DURABLE AND REACTIVE REGENERABLE MAGNESIUM-BASED SORBENTS FOR CO2 SEPARATION IN COAL GASIFICATION PROCESS**

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1. ABSTRACT

Program Introduction: Rationale and Objective

Separation of CO_2 from coal derived synthesis gas for capture and sequestration is a key technology in the reduction of greenhouse gas emissions to the environment. Existing technologies can be applied to capture CO_2; however, such applications require expensive solvents and operate at relatively low temperatures(i.e., T<40°C), imparting a severe energy penalty on the system. Therefore, advanced high temperature processes that are based on dry regenerable sorbents offer attractive advantages over the existing low temperature processes. The use of alkali- or alkaline-earth metals for CO_2 capture has been previously reported in literature. Carbon dioxide can be removed from raw coal gas by magnesium-based sorbents in the temperature range of 350°-550°C at 10 to 50 bar pressure. These temperature and pressure ranges closely match those expected in IGCC processes, including high temperature water-gas-shift operation.

In the magnesium oxide-based cyclic process, CO_2 absorption is accomplished through carbonation of magnesium oxide at IGCC process condition, while sorbent regeneration is carried out by decomposition of the carbonate either by decreasing the pressure or increasing the temperature and produces a concentrated stream of CO_2. However, to make such regenerative processes economically viable, highly durable, reactive, and attrition resistant sorbents are required to minimize attrition losses and the sorbent make-up rate.

The overall objective of this program is to develop MgO-based sorbents with chemical and physical characteristics and long term durability that permit cyclic regeneration over many cycles with very low fresh sorbent make-up rate for fluidized bed or moving bed applications.

Accomplishment Achieved During the Current Period of Performance

During the current period of performance, over twenty (20) different formulations of magnesium-based sorbents were prepared by various applications of the sol-gel technique. The sorbents were prepared in both spherical

pellets (d.=2-3 mm) and granules (d=100-300 μm) forms. One set of sorbent formulations was prepared by impregnation of calcined alumina pellets and granules (prepared by the sol-gel technique) using magnesium salts and the second set of sorbents was prepared by incorporation of magnesium salt during the gelation process. An oil drop granulation technique was employed in shaping the gel (alumina or alumina-magnesium mix) into spherical pellets of various diameters. The sorbent granules were produced by drying, crushing, and screening the gel. All pellets and granules were dried at 40ºC and calcined at 550ºC.

The impregnated sorbent formulations were prepared using 2M magnesium salt solution. All impregnated pellets appeared to be physically strong. The BET surface areas of the pellets ranged from 161 to 195 m^2/g. The magnesium content of the sorbents was about 6%. The sorbent formulations in pellet form were also prepared by incorporation of magnesium salt into the alumina sol prior to gelation. The magnesium content of the sorbent was about 10%. The sorbents prepared by incorporation of magnesium salt in the gel had much higher BET surface area (i.e., 282 compared to 161-195 m^2/g), but appear to lack sufficient physical strength. Four (4) sorbent formulations in granular form (d=100-300 μm) were also prepared by incorporation of magnesium salt into the alumina sol prior to gelation. The magnesium content of the sorbents ranged from 10% to 20%. These granular sorbents appear to have very high physical strength and good BET surface areas (i.e., 120-142 m^2/g). The attrition resistance of one of the formulations (i.e., EC19-B5) was determined according to ASTM-5757 method and the result indicates that the attrition index of the sorbent is about 0.04%, which is 100 times lower than the minimum required (i.e., 4%) for the fluidized bed applications.

The reactivities of the sorbent toward CO_2 at atmospheric pressure were determined in a thermogravimetric Analyzer (TGA) unit. The results suggest that, at the low CO_2 partial pressures used in the experiments, the reactivity of the sorbent toward CO_2 was very low. Potassium salts were added to the sorbent formulations to enhance the reactivity of the sorbent and the sorbents were evaluated in the TGA unit. The results of these tests indicated marginal improvement in the reactivity of the sorbent toward CO_2 at atmospheric pressure.

A new high pressure packed/fluidized bed was designed and is currently under construction to evaluated the sorbents performance at elevated pressures.

Plans for the Remaining Period of Performance

The work planned for the remaining month of the Phase-I of this research program includes the following tasks:

1. Evaluation of the Sorbents at Elevated Pressures. Each sorbent formulation will be subjected to 1½ cycles of absorption /regeneration in high pressure Packed-/Fluid-bed reactor. These tests will be carried out using different CO_2 partial pressure to promote either absorption or regeneraion during the respective ½ cycles. The effect of temperature on the performance of the sorbents will also be addressed. Based on the results of these tests the "best" sorbent formulations and the optimum operating conditions will be identified for durability tests
2. Sorbent Durability Tests. These tests will be carried out with the "best" sorbent at the optimum operating condition in a high pressure TGA unit to determine the effect of repeated cycling (about 10 cycles) on sorbent performance.

2. LIST OF PUBLISHED JOURNAL ARTICLES, COMPLETED PRESENTATIONS AND STUDENTS RECEIVING SUPPORT FROM THE GRANT

Conference Presentations

None. A paper will be prepared for presentation for presentation at the AIChE annual meeting in Fall 2004.

Student Supported Under this Contract

Armin H. Khayyat, PhD candidate in the department of Chemical and Environmental Engineering at The Illinois Institute of Technology.